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Unrecognized Cycloaddition Reactions of N-Alkyl-α-β-Unsaturated Imines Occurring in Biosystems and Their Biological Roles

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Abstract

© 2016, Springer Science+Business Media New York.Despite being fundamentally ubiquitous and important species in organic and bioorganic chemistry, the reactivities of N-alkyl-α-β-unsaturated imines have not been thoroughly explored due to their instability. Here, we describe novel reactivities of N-alkyl-α,β-unsaturated imines, which derived from α,β-unsaturated aldehydes (e.g., unsubstituted or substituted acrolein), alkylamines (e.g., amino-alcohols or diamines), and formaldehyde to produce 1,5-diazacyclooctanes, hexahydropyrimidines, and 1,3,5-triazacyclooctanes via formal [4 + 4], [4 + 2], and [4 + 2 + 2] cycloadditions in a stereocontrolled manner. We then also synthetically demonstrated that reaction between acrolein and biogenic amines (e.g., polyamines, noradrenaline, sphingosine), which ubiquitously exist in biosystems, proceed smoothly to give the corresponding 1,5-diazacyclooctanes. Finally, we also examined biological functions of the cycloaddition products and revealed for the first time their roles in oxidative stress mechanism and inhibition of amyloid-β (Aβ) 1-40 fibrillization.

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Keywords

1,3,5-Triazacyclooctane, 1,5-Diazacyclooctane, Hexahydropyrimidine, Imino cycloaddition, N-Alkyl-α,β-unsaturated imine